533 Rec'd PCT/PTO 27 SEP 2001

FORM PF(*-1390 U.S. DEPARTMENT OF COMN (REV. 11-2000)	SERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER	
TRANSMITTAL LETTER	TO THE UNITED STATES	Mo-6633/LeA 33,661	
DESIGNATED/ELECTE	ED OFFICE (DO/EO/US)	US APPLICATION NO (If known, see 37 CFR 1 5	
CONCERNING A FILING		To Be Assigned 37 9 9 9	
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED	
PCT/EP00/02385	17 March 2000 (17.03.00)	30 March 1999 (30.03.99)	
TITLE OF INVENTION ERASABLE OP			
APPLICANT(S) FOR DO/EO/US BERNE	TH, Horst; BIERINGER, Thomas; HAG	GEN, Rainer and KOSTROMINE,	
0 '			
Applicant herewith submits to the United Sta		the following terms and other visits	
1. X This is a FIRST submission of items			
2. This is a SECOND or SUBSEQUEN	iT submission of items concerning a filing	under 35 U.S.C. 3/1.	
3. X This is an express request to begin r items (5), (6), (9) and (21) indicated	national examination procedures (35 U.S.C.	371(f)). The submission must include	
4. X The US has been elected by the expi	ration of 19 months from the priority date (Article 31).	
5. X A copy of the International Applicati	on as filed (35 U.S.C. 371(c)(2))		
	d only if not communicated by the Internati	ional Bureau).	
b. has been communicated by	the International Bureau.	wing Office (PO/US)	
345	ication was filed in the United States Received		
6. An English language translation of the	he International Application as filed (35 U.S.	3.C. 3/1(C)(2)).	
a. X is attached hereto.	itted under 35 U.S.C. 154(d)(4).		
b. has been previously submit	ternational Aplication under PCT Article 19	(35 U.S.C. 371(c)(3))	
a. are attached hereto (requir	red only if not communicated by the Interna		
An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). a.			
c. have not been made; however, the time limit for making such amendments has NOT expired.			
c. have not been made; however, the time limit for making such amendments has NOT expired. d. have not been made and will not be made.			
8. An English language translation of t	the amendments to the claims under PCT At	rticle 19 (35 U.S.C. 371 (c)(3)).	
9 X An oath or declaration of the inven			
1	the annexes of the International Preliminary	Examination Report under PCT	
Article 36 (35 U.S.C. 371(c)(5)).			
Items 11 to 20 below concern docume	ent(s) or information included:		
1	nent under 37 CFR 1.97 and 1.98.		
1	ording. A separate cover sheet in compliance	be with 37 CFR 3.28 and 3.31 is included.	
	preminary amendment.		
15. A substitute specification.	- 1/ 11 - 1-1-1		
16. A change of power of attorney a		1 12 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	
17. A computer-readable form of the	sequence listing in accordance with PCT R	ule 13ter.2 and 35 U.S.C. 1.621 - 1.623.	
18. A second copy of the published	international application under 35 U.S.C. 15	54(d)(4).	
19. A second copy of the English la	inguage translation of the international appli	cation under 35 U.S.C. 154(d)(4).	
20. X Other items or information:			
Form PTO 1449 w/references			
Abstract			
Drawing (1 sheet)			

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21. X The follow					CAI	CULATIONS P	TO USE ONLY
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):							
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a (2)) paid to USPTO and International Search Report not prepared by the EPO or JPO			\$1000 .0 0				
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$860.00							
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO							
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00							
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)							
					\$	860.00	
Surcharge of \$130.00 months from the ear	liest claimed prior	ity date (37 Cl	FR 1.492(e)).	20 30	\$	0.00	
CLAIMS	NUMBER FIL		MBER EXTRA	RATE	\$		
Total claims	14 -20		0	x \$18.00	\$	0.00	
Independent claims MULTIPLE DEPEN	1 -3		0	x \$80.00	\$	0.00	
MUGHTLE DEFEN				+ \$270.00	\$	0.00	
Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above				\$	860.00		
			Si	UBTOTAL =	\$	860.00	*
Processing fee of \$130.00 for furnishing the English translation later than 20 30 months from the earliest claimed priority date (37 CFR 1.492(f)).			\$	0.00			
TOTAL NATIONAL FEE =			NAL FEE =	\$	860.00		
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property			\$	40.00			
TOTAL FEES ENCLOSED =			\$	900.00			
- 3-4 - 3-4 - 3-3 - 3-3						ount to be refunded:	\$
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b. 🔀 Please cha A duplicat	rge my Deposit A e copy of this shee	ccount No. 17	3-3848 ii	he above fees is enclo	0.00		
c. X The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3848. A duplicate copy of this sheet is enclosed.							
d. Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.							
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.							
SEND ALL CORRESPONDENCE TO Jones A. Wishe							
SIGNATURE							
				James F	R. Fra	nks	***
				42,552			
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PATENT APPLICATION Mo-6633 LeA 33,661

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATION OF)) PCT/EP00/02385
HORST BERNETH ET AL)
SERIAL NUMBER: TO BE ASSIGNED	,))
FILED: HEREWITH	,))
TITLE: ERASABLE OPTICAL RECORDING MATERIAL FOR BLUE LASERS	,)))

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents Washington, D.C. 20231

Sir:

This preliminary amendment is being filed concurrently with the subject patent application. Upon granting a Serial Number and filing date, please amend the subject patent application as follows.

"Express Mail" mailing l	abel number	ET14	6893758US	
Date of Deposit	September	27,	2001	
I hereby certify that the Postal Service "Expresa 1.10 on the date Indicator of Patents and Trademontal Patents and Trademonta	ss Mail Post Office to ted above and is add	to Addr Iressed	essee" service un to the Assistant C	der 37 CFR
I	onna J. Ve	atch		
(),	(Name of person mail)	ng paper	or tee)	

Signature of person mailing paper or fee)

Please amend the Application as follows.

IN THE ABSTRACT:

Please add the following on a separate page.

-- ERASABLE OPTICAL RECORDING MATERIAL FOR BLUE LASERS

ABSTRACT OF THE DISCLOSURE

An optical recording material for binary, multibit or volume data storage is described. The optical recording material comprises: (a) at least one dyestuff selected from polymeric azo dyestuffs and oligomeric azo dyestuffs, the dyestuff changing its spatial arrangement upon irradiation with polarized electromagnetic radiation; and (b) optionally at least one grouping having form anisotropy. The optical recording material has the following characteristics: (i) the absorption maximum of the dyestuff(s) is at least 30 nm less than 400 nm and/or at least 30 nm greater than 400 nm; (ii) at 400 nm the dyestuff reaches an optical density of not more than 60% of its absorption maximum; (iii) the optical recording material has the capacity for being rewritten on by changing the state of polarization of actinic light, an intensity of at least 80% of the original value being achieved after a deletion/rewriting cycle; and (iv) at 400 nm, under identical conditions, an optical writing operation performed upon the optical recording material proceeds no more slowly than at 500 nm, and birefringence values induced during the optical writing operation do not differ from those birefringence values induced at 500 nm by more than 10% .--

An abstract is included herewith on a separate page.

IN THE SPECIFICATION:

Please replace the title at line 1 of page 1 with the following.

-- ERASABLE OPTICAL RECORDING MATERIAL FOR BLUE LASERS--

Please insert the following between lines 1 and 3 on page 1 of the specification.

Mo-6633

-- CROSS REFERENCE TO RELATED PATENT APPLICATIONS

The present patent application claims the right of priority under 35 U.S.C. 119 (a)-(d) and 35 U.S.C. 365 of International Application No. PCT/EP00/02385, filed 17 March 2000, which was published in German as International Patent Publication No. WO 00/60586 on 12 October 2000, which is entitled to the right of priority of German Patent Application No. 199 14 325.0, filed 30 March 1999.

FIELD OF THE INVENTION --

Please insert the following at line 6 on page 1 of the specification.
--BACKGROUND OF THE INVENTION--

Please insert the following at line 27 on page 2 of the specification.
--SUMMARY OF THE INVENTION--

Please insert the following between lines 24 and 26 on page 3 of the specification.

-- BRIEF DESCRIPTION OF THE DRAWING

Figure 1 is a graphical representation of Δn (birefringence) as a function of time during repetitive writing / deletion experiments performed on an optical recording material according to the present invention, as represented by Example 3 herein.

DETAILED DESCRIPTION OF THE INVENTION--

Please replace line 1 of page 30 of the specification with the following. WHAT IS CLAIMED IS:

IN THE CLAIMS:

Please replace Claims 1-14 with the following

1. (Once Amended, Clean) An optical recording material for at least one of Mo-6633

binary, multibit and volume data storage, comprising:

- at least one dyestuff selected from polymeric azo dyestuffs and oligomeric azo dyestuffs, said dyestuff changing its spatial arrangement upon irradiation with polarized electromagnetic radiation; and
- (b) optionally at least one grouping having form anisotropy, wherein
- (i) the absorption maximum of the dyestuff is at least one of, at least 30 nm less than 400 nm and at least 30 nm greater than 400 nm,
- (ii) at 400 nm the dyestuff reaches an optical density of not more than 60% of its absorption maximum,
- (iii) said optical recording material has the capacity for being rewritten on by changing the state of polarization of actinic light, an intensity of at least 80% of the original value being achieved after a deletion/rewriting cycle, and
- (iv) wherein at 400 nm, under identical conditions, an optical writing operation upon said optical recording material proceeds no more slowly than at 500 nm, and birefringence values induced during said optical writing operation do not differ from those birefringence values induced at 500 nm by more than 10%.
- 2. (Once Amended, Clean) The recording material of Claim 1 wherein the absorption maximum of the dyestuff is less than 370 nm.
- 3. (Once Amended, Clean) The recording material of Claim 1 wherein the absorption maximum of the dyestuff is greater than 450 nm.
- 4. (Once Amended, Clean) The recording material of Claim 1 wherein it comprises a copolymer which comprises at least one component of which the

absorption maximum is greater than 450 nm, and at least one component of which the absorption maximum is less than 360 nm.

- 5. (Once Amended, Clean) The recording material of Claim 1 wherein in the solid state at a thickness of 250 nm said recording material has an optical density of ≤ 1, at a wavelength in a wavelength range from 380 to 420 nm.
- 6. (Once Amended, Clean) The recording material of Claim 1 wherein said optical recording material is optically written upon using electromagnetic radiation that is light in a laser wavelength range of between 380 to 420 nm.
- 7. (Once Amended, Clean) The recording material of Claim 1 wherein the dyestuff corresponds to the formula (I):

$$X^{1}$$
 $(R^{1})_{m}$
 $(R^{2})_{n}$
 $(I),$

wherein

R¹ and R² independently of one another represent hydrogen or a nonionic substituent,

m and n independently of one another represent an integer from 0 to 4,

 X^1 and X^2 denote $X^{1'}$ - R^3 or $X^{2'}$ - R^4 ,

X¹' and X²' represent a direct bond, -O-, -S-, -(N-R⁵)-, -C(R⁶R⁻)-, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁶)- or -(CNR⁶-NR⁵)-,

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R³, R⁴, R⁵ and R⁸ independently of one another represent hydrogen,

$$\begin{split} &C_{1^-} \text{ to } C_{20}\text{-alkyl, } C_{3^-} \text{ to } C_{10}\text{-cycloalkyl, } C_{2^-} \text{ to } C_{20}\text{-alkenyl, } C_{6^-} \text{ to } \\ &C_{10}\text{-aryl, } C_{1^-} \text{ to } C_{20}\text{-alkyl-}(C=O)\text{-, } C_{3^-} \text{ to } C_{10}\text{-cycloalkyl-}(C=O)\text{-, } C_{2^-} \\ &\text{to } C_{20}\text{-alkenyl-}(C=O)\text{-, } C_{6^-} \text{ to } C_{10}\text{-aryl-}(C=O)\text{-, } C_{1^-} \text{ to } C_{20}\text{-alkyl-} \\ &(SO_2)\text{-, } C_{3^-} \text{ to } C_{10}\text{-cycloalkyl-}(SO_2)\text{-, } C_{2^-} \text{ to } C_{20}\text{-alkenyl-}(SO_2)\text{- or } \\ &C_{6^-} \text{ to } C_{10}\text{-aryl-}(SO_2)\text{-, } \end{split}$$

X¹-R³ and X²-R⁴ can represent hydrogen, halogen, cyano, nitro, CF₃ or CCl₃,

 R^6 and R^7 independently of one another represent hydrogen, halogen, C_1 -to C_{20} -alkyl, C_1 - to C_{20} -alkoxy, C_3 - to C_{10} -cycloalkyl, C_2 - to C_{20} -alkenyl or C_6 - to C_{10} -aryl,

wherein the sensitivity of the dyestuff to actinic light after induced birefringence has been written in is retained at a final value, and this sensitivity is at least 5% of the original value when the longitudinal axis of the molecule lies perpendicular to the direction of polarization of the actinic light.

8. (Once Amended, Clean) The recording material of Claim 1 wherein said recording material is prepared from at least one monomer of the formula (II),

wherein

- R represents hydrogen or methyl,
- S¹ represents a direct bond, -O-, -S- or -NR⁹-,
- T¹ represents -(CH₂)_p-, where the chain can be interrupted by -O-, -NR⁹- or -OSiR¹⁰₂O-,
- Q¹ represents -O-, -S-, -(N-R⁵)-, -C(R⁶R⁻)-, -(C=O)-, -(CO-O)-, -(CO-NR⁶)-, -(SO₂)-, -(SO₂-O)-, -(SO₂-NR⁶)-, -(C=NRឹ)-, -(CNRឹ-NR⁶)-, -(CH₂)p-, p- or m-C₆H₄- or a divalent radical of the formula

- represents an integer from 0 to 4, where for i > 1 the individual Q¹ can have different meanings,
- X^2 represents $X^{1'}$ - R^3 or $X^{2'}$ - R^4 ,
- $X^{1'}$ and $X^{2'}$ each independently represent a direct bond, -O-, -S-, -(N-R⁵)-, $C(R^6R^7)$ -, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, -(SO₂)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁸)- or -(CNR⁸-NR⁵)-,

R¹ and R² each independently represent hydrogen or a nonionic substituent,

R³ and R⁴ each independently represent hydrogen, C_1 - to C_{20} -alkyl, C_3 - to C_{10} -cycloalkyl, C_2 - to C_{20} -alkenyl, C_6 - to C_{10} -aryl, C_1 - to C_{20} -alkyl-(C=O)-, C_3 - to C_{10} -cycloalkyl-(C=O)-, C_2 - to C_{20} -alkenyl-(C=O)-, C_6 - to C_{10} -aryl- (C=O)-, C_1 - to C_{20} -alkyl-(SO₂)-, C_3 - to C_{10} -cycloalkyl-(SO₂)-, C_2 - to C_{20} - alkenyl-(SO₂)- or C_6 - to C_{10} -aryl-(SO₂)-, and

m and n each independently represent an integer from 0 to 4.

9. (Once Amended, Clean) The recording material of Claim 1 wherein it comprises at least one polymer selected from formulas XIII to XX,

$$\begin{array}{c|c} & & & \\ & & \\ & & & \\ & & & \\ & & & \\ & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ &$$

$$\begin{array}{c|c} & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & \\ & & & \\ & \\ & & \\ & & \\ & & \\ & & \\ & & \\ & & \\ & & \\ & & \\ & & \\ & & \\ & & \\ & & \\ &$$

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wherein the compositions in the co- and terpolymers can vary, provided that x + y adds up to 100 mol %, or x + y + z adds up to 100 mol %.

10. (Once Amended, Clean) The recording material of Claim 9 wherein p is between 10 and 1,000,

in the case of the copolymers, the molar ratio of x : y is between 10:90 and 90:10, and

in the case of terpolymers, x + y is greater than 10 mol%.

- 11. (Once Amended, Clean) A storage system comprising the recording material of Claim 1.
- 12. (Once Amended, Clean) The storage system of Claim 11 wherein the recording material comprises one or more unsupported objects of any desired shape.
- 13. (Once Amended, Clean) The storage system of Claim 11 wherein it also additionally comprises a reflection layer.
- 14. (Once Amended, Clean) A process for the production of the storage system of Claim 11 wherein said process comprises applying the storage medium by spin-coating.

REMARKS

Claims in the case are 1-14. Claims 1-14 have been amended herein as to form, for example, by introducing indentation, indefinite and definite articles, and replacing multiple dependencies with single dependencies. Basis for the additions to Claim 8 herein (e.g., S¹, T¹ etc.) are found in Claim 7, and at page 9, lines 6-20 of the specification.

The specification has been amended to include section headings, in accordance with accepted practice before the Office. Basis for the Brief Description of the Drawing inserted herein on page 3 of the specification is found at page 27, lines 5-16, and page 28, lines 12-19 of the specification. The title has been changed to correspond to that of the related International Patent Publication No. WO 00/60586. Page 1 of the application has been amended herein to introduce cross reference information. The cross reference information is presented in accordance with 37 C.F.R. 1.78(a)(2) (Federal Register / Vol. 65, No. 183 / Wednesday, September 20, 2000; Changes to Implement Eighteen-Month Publication of Patent Applications; Final Rule). An abstract of the disclosure is also included herewith on a separate page.

The amendments presented herein are not believed to represent the entry of new matter into the application. Applicants respectfully request entry of this preliminary amendment.

Respectfully submitted,

James R. Franks
Agent for Applicants

Reg. No. 42,552

Bayer Corporation 100 Bayer Road Pittsburgh, Pennsylvania 15205-9741 (412) 777-8339 FACSIMILE PHONE NUMBER: (412) 777-8363 /vjt/JRF0089

VERSIONS WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION: (Marked-Up)

The following are changes and additions made to the specification.

The following is a version of the title of the application on line 1 of page 1, showing changes made thereto herein.

[Optical recording material for blue lasers which can be rewritten on]

ERASABLE OPTICAL RECORDING MATERIAL FOR BLUE LASERS

Line 1 of page 30 of the specification has been amended as follows. [Patent claims] WHAT IS CLAIMED IS:

IN THE CLAIMS: (Marked-Up)

The following are versions of the amended claims with markings to show changes made thereto in the present Preliminary Amendment.

- 1. (Once Amended, Marked-Up) An [O]optical recording material for at least one of binary, [and/or] multibit and[/or] volume data storage, comprising:
 - (a) at least one <u>dyestuff selected from</u> polymeric <u>azo dyestuffs</u> and[/or] oligomeric azo dyestuff<u>s</u>, [which] <u>said dyestuff</u> chang[es]<u>ing</u> its spatial arrangement <u>up</u>on irradiation with polarized electromagnetic radiation[,]; and
 - (b) optionally at least one grouping having form anisotropy, [characterized in that]

wherein

[-] (i) the absorption maximum of the dyestuff is at least one of, at least 30 nm[, preferably 40 nm, different from] less than 400 nm and at least 30 nm greater than 400 nm, [and]

Mo-6633

- [-] (ii) at 400 nm the dyestuff reaches an optical density of not more than 60% of its absorption maximum, [and]
- [-] (iii) [there is] said optical recording material has the capacity for being rewritten on by changing the state of polarization of [the] actinic light, an intensity of at least 80% of the original value being achieved after a deletion/rewriting cycle, and
- [-] (iv) wherein at 400 nm, under [otherwise] identical conditions, [the] an optical writing operation upon said optical recording material proceeds no more slowly than at 500 nm, and birefringence values induced [here] during said optical writing operation do not differ from those birefringence values induced at 500 nm by more than 10%.
- 2. (Once Amended, Marked-Up) <u>The [R]recording material [according to claim 1, characterized in that] of Claim 1 wherein the absorption maximum [(AM)] of the dyestuff is less than 370 nm[, preferably 360 nm].</u>
- 3. (Once Amended, Marked-Up) <u>The [R]recording material [according to claim 1, characterized in that] of Claim 1 wherein the absorption maximum of the dvestuff is greater than 450 nm.</u>
- 4. (Once Amended, Marked-Up) <u>The [R]recording material [according to claim 1, characterized in that] of Claim 1 wherein it comprises a copolymer which comprises at least one component of which the [AM] <u>absorption maximum</u> is greater than 450 nm, and at least one component of which the [AM] <u>absorption maximum</u> is less than 360 nm.</u>
- 5. (Once Amended, Marked-Up) The [R]recording material [according to one or more of claims 1 to 4, characterized in that] of Claim 1 wherein in the solid state at a thickness of 250 nm [it] said recording material has an optical density of ≤ 1 , Mo-6633

[preferably less than or equal to 0.5, especially preferably less than or equal to 0.3,] at a wavelength in a wavelength range from 380 to 420 nm[, preferably 390 to 410 nm, especially preferably 395 to 405 nm].

- 6. (Once Amended, Marked-Up) The [R]recording material [according to one or more of claims 1 to 5, characterized in that] of Claim 1 wherein said optical recording material is optically written upon using [the] electromagnetic radiation that is light in [the] a laser wavelength range of [preferably] between 380 to 420 nm[, particularly preferably between 390 and 410 nm, especially preferably between 395 and 405 nm].
- 7. (Once Amended, Marked-Up) <u>The [R]recording material [according to any one of claims 1 to 6, characterized in that] of Claim 1 wherein the [chemically bonded] dyestuff corresponds to the formula (I):</u>

$$X^1$$
 $(R^1)_m$
 (I) ,
 $(R^2)_m$

wherein

R¹ and R² independently of one another represent hydrogen or a nonionic substituent, [and]

m and n independently of one another represent an integer from 0 to 4[, preferably 0 to 2, where],

 X^1 and X^2 denote $X^{1'}$ - R^3 or $X^{2'}$ - R^4 , [and]

R³, R⁴, R⁵ and R⁵ independently of one another represent hydrogen, $C_{1^-} \text{ to } C_{20^-} \text{alkyl}, \ C_{3^-} \text{ to } C_{10^-} \text{cycloalkyl}, \ C_{2^-} \text{ to } C_{20^-} \text{alkenyl}, \ C_{6^-} \text{ to } C_{10^-} \text{aryl}, \ C_{1^-} \text{ to } C_{20^-} \text{alkyl-} (C=O)_-, \ C_{3^-} \text{ to } C_{10^-} \text{cycloalkyl-} (C=O)_-, \ C_{2^-} \text{ to } C_{20^-} \text{alkenyl-} (C=O)_-, \ C_{1^-} \text{ to } C_{20^-} \text{alkyl-} (SO_2)_-, \ C_{3^-} \text{ to } C_{10^-} \text{cycloalkyl-} (SO_2)_-, \ C_{2^-} \text{ to } C_{20^-} \text{alkenyl-} (SO_2)_- \text{ or } C_{6^-} \text{ to } C_{10^-} \text{aryl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{alkenyl-} (SO_2)_- \text{ or } C_{1^-} \text{ to } C_{20^-} \text{alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{alkenyl-} (SO_2)_- \text{ or } C_{1^-} \text{ to } C_{20^-} \text{alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ to } C_{20^-} \text{alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_{1^-} \text{ to } C_{20^-} \text{ alkenyl-} (SO_2)_-, \ C_$

X1-R3 and X2-R4 can represent hydrogen, halogen, cyano, nitro, CF3 or CCl3,

R⁶ and R⁷ independently of one another represent hydrogen, halogen, C_1 -to C_{20} -alkyl, C_1 - to C_{20} -alkoxy, C_3 - to C_{10} -cycloalkyl, C_2 - to C_{20} -alkenyl or C_6 - to C_{10} -aryl[.],

wherein the sensitivity of the dyestuff to actinic light after [the] induced birefringence has been written in is retained at a final value, and this sensitivity is at least 5%[, preferably 10%, particularly preferably 15%, especially preferably 20%] of the original value when the longitudinal axis of the molecule lies perpendicular to the direction of polarization of the actinic light.

8. (Once Amended, Marked-Up) <u>The [R]recording material [according to any one of claims 1 to 7, characterized in that] of Claim 1 wherein said recording material is prepared from at least one monomer of the formula (II).</u>

wherein

R represents hydrogen or methyl, [and]

[the other radicals have the above-mentioned meaning,

has been used in the preparation]

- S¹ represents a direct bond, -O-, -S- or -NR⁹-,
- T¹ represents -(CH₂)₂-, where the chain can be interrupted by -O-, -NR⁹or -OSiR¹⁰₂O-,
- Q¹ represents -O-, -S-, -(N-R⁵)-, -C(R⁶R⁻)-, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, $-(SO_2)-, -(SO_2-O)-, -(SO_2-NR⁵)-, -(C=NR⁶)-, -(CNR⁶-NR⁵)-, -(CH_2)_p-, p- or \\ -C_6H_4- or a divalent radical of the formula$

- i represents an integer from 0 to 4, where for i > 1 the individual Q¹ can have different meanings.
- X^2 represents X^1-R^3 or X^2-R^4 ,

X¹¹ and X² each independently represent a direct bond, -O-, -S-, -(N-R⁵)-, - $C(R^6R^7)$ -, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, -(SO₂)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁶)- or -(CNR⁶-NR⁵)-,

R¹ and R² each independently represent hydrogen or a nonionic substituent, R³ and R⁴ each independently represent hydrogen, C_1 - to C_{20} -alkyl, C_3 - to C_{10} -cycloalkyl, C_2 - to C_{20} -alkenyl, C_6 - to C_{10} -aryl, C_1 - to C_{20} -alkyl-(C=O)-, C_3 - to C_{10} -cycloalkyl-(C=O)-, C_2 - to C_{20} -alkenyl-(C=O)-, C_6 - to C_{10} -aryl-(C=O)-, C_1 - to C_{20} -alkyl-(SO₂)-, C_3 - to C_{10} -cycloalkyl-(SO₂)-, C_2 - to C_{20} -alkenyl-(SO₂)-, and

m and n each independently represent an integer from 0 to 4.

9. (Once Amended, Marked-Up) The [R]recording material [according to any one of claims 1 to 8, characterized in that] of Claim 1 wherein it comprises at least one [of the] polymer[s of the] selected from formulas XIII to XX.

$$\begin{array}{c|c} & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & \\ & & & \\ & &$$

[e.g. x=22 mol%]

[e.g. y=30 mol%]

[e.g. z=48 mol%]

$$\begin{array}{c|c} & & & \\ & & \\ & & & \\ & & \\ & & \\ & & & \\ & & \\ & & & \\ & & \\ & & \\ & & & \\ & & \\ & & & \\ & &$$

$$\begin{array}{c|c} & & & \\ & & \\ & & & \\ & & \\ & & \\ & & & \\ & & \\ & & & \\ & & \\ & & \\ & & & \\ & & \\ & & & \\ & &$$

wherein the compositions in the co- and terpolymers can vary, provided that x + y adds up to 100 mol%, or x + y + z adds up to 100 mol%.

10. (Once Amended, Marked-Up) <u>The [R]recording material [according to claim 9, characterized in that] of Claim 9 wherein</u>

p is between 10 and 1,000, [and/or]

in the case of the copolymers, the molar ratio of x: y is between 10:90 and 90:10, [preferably between 30:70, particularly preferably between 40:60 and 60:40, and especially preferably 50:50,] and[/or]

in the case of terpolymers, x + y is greater than 10 mol%[, preferably greater than 20 mol%, particularly preferably greater than 30 mol%].

- 11. (Once Amended, Marked-Up) A [S]storage system[, characterized in that it] compris[es]ing [a] the recording material [according to any of claims 1 to 10] of Claim 1.
- 12. (Once Amended, Marked-Up) The [S]storage system [according to claim 11, characterized in that] of Claim 11 wherein the recording material comprises one or more unsupported objects of any desired shape[, preferably an unsupported flat structure, particularly preferably an unsupported film, a multi-layer build-up preferably comprising at least one substrate layer].
- 13. (Once Amended, Marked-Up) The [S]storage system [according to claim 11, characterized in that] of Claim 11 wherein it also additionally comprises a reflection layer.
- 14. (Once Amended, Marked-Up) A [P]process for the production of the storage system [according to at least one of claims 11 or 12, or 13, characterized in that it] of Claim 11 wherein said process comprises [a step in which] applying the storage medium [is applied] by spin-coating.

ERASABLE OPTICAL RECORDING MATERIAL FOR BLUE LASERS

ABSTRACT OF THE DISCLOSURE

An optical recording material for binary, multibit or volume data storage is described. The optical recording material comprises: (a) at least one dyestuff selected from polymeric azo dyestuffs and oligomeric azo dyestuffs, the dyestuff changing its spatial arrangement upon irradiation with polarized electromagnetic radiation; and (b) optionally at least one grouping having form anisotropy. The optical recording material has the following characteristics: (i) the absorption maximum of the dyestuff(s) is at least 30 nm less than 400 nm and/or at least 30 nm greater than 400 nm; (ii) at 400 nm the dyestuff reaches an optical density of not more than 60% of its absorption maximum; (iii) the optical recording material has the capacity for being rewritten on by changing the state of polarization of actinic light, an intensity of at least 80% of the original value being achieved after a deletion/rewriting cycle; and (iv) at 400 nm, under identical conditions, an optical writing operation performed upon the optical recording material proceeds no more slowly than at 500 nm, and birefringence values induced during the optical writing operation do not differ from those birefringence values induced at 500 nm by more than 10%.

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Optical recording mate	rial for blue lasers which can be rewritten 1937999

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The present invention relates to an optical recording material for binary and/or multibit and/or volume data storage, the preparation thereof and its use as a storage material at wavelengths of about 400 nm.

Materials for binary and/or multibit and/or volume data storage which have azo dyestuffs as antennae for the incident light already exist in the prior art. In particular, the class of side chain polymers in which, in addition to the azo dyestuffs, components having form anisotropy are also used as side chains, is distinguished by the level of the birefringence which can be induced by light. In these materials, the dyestuffs, in combination with components having form anisotropy, bring about an orientation in the actinic light field if they are irradiated with polarized light of a suitable wavelength: A light-induced birefringence builds up.

US Patent US 5 384 221 describes an optical recording material for binary and/or multibit and/or volume data storage comprising at least one polymeric and/or oligomeric azo dyestuff which changes its spatial arrangement on irradiation with polarised electromagnetic radiation, and optionally at least one grouping having form anisotropy.

However, these materials do not meet all requirements, and in particular, because of their absorption properties (too high an optical density) at 400 nm, they cannot be used for data storage: In reflection constructions, the actinic light is already noticeably absorbed in the recording layers after a few layers of atoms, and the actinic light cannot act on the complete thickness of the recording material. This means on the one hand that during storage of optical information a significant increase in the sample temperature occurs due to the absorption of light. A significant increase in the sample temperature is understood as meaning a temperature increase of at least 20°C, more precisely of at least 50°C, in particular of

more than 100°C, and especially particularly of at least 140°C. This increase in temperature can be measured, for example, with an IR thermal head (Inframetrics Thermocam PM 290).

On the other hand, the high absorption impedes reading of the light-induced information with the wavelengths which have been used for writing: The light arriving from the recording material at the detection construction is too weak still to have an adequate signal/noise ratio. An adequate signal/noise ratio exists when the intensity of the detected light stands out from the scatter background by an order of magnitude.

These known materials sometimes do not show an adequate capacity for being rewritten on: It has already been shown that in the case of irradiation with linearly polarized light the previously induced birefringence can be deleted by rotating the direction of polarization of the actinic light by, for example, 90°. Alternatively, the birefringence can be reduced, for example, by using circularly polarized light. During a renewed writing operation, however, on irradiation with linearly polarized light, in the prior art the birefringence value such as has been achieved during writing on an isotropic sample is not produced.

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For the abovementioned reasons (too high an optical density), rewriting on was not possible, especially at wavelengths of about 400 nm.

The capacity for being written on at 400 nm is of great importance, however, for the use of the blue laser diodes (NICHIA) which have just become commercially obtainable, since these laser diodes allow the generation of high storage densities.

There was accordingly the need for a recording material which can be used for optical data storage for the wavelength range of blue laser diodes. This material should achieve in puncto light-inducible birefringence of comparable values on

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exposure in the blue spectral range (about 400 nm) to those on exposure in the green spectral range (about 515 nm). It should furthermore have a capacity for being rewritten on.

It has been found, surprisingly, that the recording materials described in this application meet the stated requirements.

The invention accordingly provides an optical recording material which can be employed for binary and/or multibit and/or volume data storage and comprises at least one polymeric and/or oligomeric azo dyestuff which changes its spatial arrangement on irradiation, and optionally at least one grouping having form anisotropy. The recording material is characterized in that

- the absorption maximum of the dyestuff is at least 30 nm, preferably 40 nm, different from 400 nm and
- at 400 nm the dyestuff reaches an optical density of not more than 60% of its absorption maximum and
- there is the capacity for being rewritten on by changing the state of polarization of the actinic light, an intensity of at least 80% of the original value being achieved after a deletion/rewriting cycle, and
- wherein at 400 nm, under otherwise identical conditions, the writing operation proceeds no more slowly than at 500 nm, and birefringence values induced here do not differ from those birefringence values induced at 500 nm by more than 10%.

This is achieved e.g. by polymers in which the absorption maximum (AM) of the dyestuff is less than 370 nm, preferably 360 nm.

However, it is also possible that polymers and/or oligomers in which the absorption maximum of the dyestuff is greater than 450 nm are used.

Very good results have been achieved if the recording material comprises a copolymer which comprises at least one component in which the AM is greater than 450 nm and at least one component in which the AM is less than 360 nm.

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Particularly advantageous effects can be achieved if the absorption band with the maximum absorption is particularly narrow.

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Furthermore, it is to be noted in particular that the recording material according to the invention has, in the solid state at a thickness of 250 nm, an optical density of ≤ 1 , preferably less than or equal to 0.5, especially preferably less than or equal to 0.3, at a wavelength in a wavelength range of 380 to 420 nm, preferably 390 to 410 nm, especially preferably 395 to 405 nm.

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The optical density can be determined with commercial UV/VIS spectrometers (e.g. CARY, 4G, UV/VIS spectrometer).

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If this is ensured, the effects according to the invention can be achieved in a favourable manner by treating the recording material with electromagnetic radiation (light) in the wavelength range of lasers of preferably between 380 to 420 nm, particularly preferably between 390 and 410 nm, especially preferably between 395 and 405 nm.

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In the most favourable case, the reading process should be carried out with light of the same wavelength. If this is not possible in some cases, the wavelength can be longer, but the wavelength should preferably be only slightly longer.

The reorientation of the dyestuff after exposure to actinic light results, for example, from polarized absorption spectroscopy studies: A sample previously exposed to actinic light is investigated between 2 polarizers in a UV/VIS spectrometer (e.g.

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CARY 4G, UV/VIS spectrometer) in the spectral range of the absorption of the dyestuffs. On rotation of the sample around the perpendicular to the sample and with a suitable position of the polarizers, for example in the crossed state, the reorientation of the dyestuff follows from the course of the intensity of the extinction as a function of the sample angle and as a result can be determined unambiguously.

An important parameter which acquires importance even more often in the following is the orientation of the longitudinal axis of the molecules. The longitudinal axis of the molecules can be determined, for example, with the aid of the molecular shape by molecular modelling (e.g. CERIUS²).

An important parameter of the dyestuffs according to the invention is their sensitivity to actinic light after the writing operation.

The holographic sensitivity, for example, is a measure of the sensitivity of a molecule to actinic light. It is calculated, for example, from the holographic growth curve, that is to say the evolution of the diffraction efficiency (= diffracted intensity with respect to the incident intensity of the reading laser) as a function of the energy deposited by the writing beams. The sensitivity is defined as the increase in the root of the diffraction efficiency according to the energy deposited, standardized to the thickness of the storage medium.

At the microscopic level, sensitivity means the probability that a photon converts a molecule from the electronic base state into an excited state, which as a rule is described with a cis-configuration.

The dyestuff decreases its absorption properties, in particular its sensitivity to actinic light, for example by folding in the direction perpendicular to the direction of polarization of the actinic light and by its longitudinal molecular axis coming to lie at an angle to the direction of polarization of the actinic light of between 10° and 90°,

preferably between 50° and 90°, particularly preferably between 75° and 90°, and especially preferably between 85° and 90°. Such dyestuffs are then no longer available for further writing operations if components of the polarizability tensor can no longer react with the actinic light, and on renewed writing after a deletion operation the birefringence value achieved during the previous writing is no longer achieved. Such dyestuffs are characterized in that, for example, their polarizability tensor has an elongated shape.

It is essential to the invention that the sensitivity to actinic light of the dyestuffs according to the invention after the induced birefringence has been written in is retained at a final value, and this sensitivity is at least 5%, preferably 10%, particularly preferably 15%, especially preferably 20% of the original value when the longitudinal axis of the molecule lies perpendicular to the direction of polarization of the actinic light.

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Rewriting on the recording material several times is possible in this way: The light-induced birefringence value achieved after the deletion operation achieves a value which differs from the previous value by at most 20%, preferably 10%, particularly preferably 5%, and especially preferably by not more than 1%.

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The capacity for being rewritten on several times is understood as meaning that at least 2, preferably at least 10, particularly preferably at least 100, and especially preferably at least 1,000 writing/deletion cycles are carried out.

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The grouping which interacts with the electromagnetic radiation is an azo dyestuff. The material according to the invention consequently comprises at least one azo dyestuff.

Azo dyestuffs have, for example, the following structure of the formula (I)

5 wherein

> independently of one another represent hydrogen or a nonionic R^1 and R^2 substituent and

independently of one another represent an integer from 0 to 4, m and n preferably 0 to 2,

denote X'-R3 or X2-R4, X1 and X2

15 wherein

> represent a direct bond, -O-, -S-, -(N-R⁵)-, -C(R⁶R⁷)-, -(C=O)-, -(CO- $X^{i'}$ and $X^{i'}$ O)-, -(CO-NR⁵)-, -(SO₂)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁸)- or -(CNR8-NR5)-,

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R3, R4, R5 and R8 independently of one another represent hydrogen, C1- to C20-alkyl, C_{3} - to C_{10} -cycloalkyl, C_{2} - to C_{20} -alkenyl, C_{6} - to C_{10} -aryl, C_{1} - to C_{20} alkyl-(C=O)-, C_3 - to C_{10} -cycloalkyl-(C=O)-, C_2 - to C_{20} -alkenyl-(C=O)-, C_6 - to C_{10} -aryl-(C=O)-, C_1 - to C_{20} -alkyl-(SO₂)-, C_3 - to C_{10} cycloalkyl-(SO₂)-, C_2 - to C_{20} -alkenyl-(SO₂)- or C_6 - to C_{10} -aryl-(SO₂)or

X¹-R³ and X²-R⁴ can represent hydrogen, halogen, cyano, nitro, CF₃ or CCl₃,

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 R^6 and R^7 independently of one another represent hydrogen, halogen, C_1 - to C_{20} -alkyl, C_1 - to C_{20} -alkoxy, C_3 - to C_{10} -cycloalkyl, C_2 - to C_{20} -alkenyl or C_6 - to C_{10} -aryl.

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- Nonionic substituents are to be understood as meaning halogen, cyano, nitro, C₁- to C₂₀-alkyl, C₁- to C₂₀-alkoxy, phenoxy, C₃- to C₁₀-cycloalkyl, C₂- to C₂₀-alkenyl or C₆- to C₁₀-aryl, C₁- to C₂₀-alkyl-(C=O)-, C₆- to C₁₀-aryl-(C=O)-, C₁- to C₂₀-alkyl-(SO₂)-, C₁- to C₂₀-alkyl-(C=O)-O-, C₁- to C₂₀-alkyl-(C=O)-NH-, C₆- to C₁₀-aryl-(C=O)-NH-, C₁- to C₂₀-alkyl-O-(C=O)-, C₁- to C₂₀-alkyl-NH-(C=O)- or C₆- to C₁₀-aryl-NH-(C=O)-.
 - The alkyl, cycloalkyl, alkenyl and aryl radicals in turn can be substituted by up to 3 radicals from the series consisting of halogen, cyano, nitro, C_1 to C_{20} -alkyl, C_1 to C_{20} -alkoxy, C_3 to C_{10} -cycloalkyl, C_2 to C_{20} -alkenyl or C_6 to C_{10} -aryl, and the alkyl and alkenyl radicals can be straight-chain or branched.
 - Halogen is to be understood as meaning fluorine, chlorine, bromine and iodine, in particular fluorine and chlorine.
- The recording material according to the invention is preferably a polymeric or oligomeric organic, amorphous material, particularly preferably a side-chain polymer.
- The main chains of the side-chain polymers originate from the following base structures: polyacrylate, polymethacrylate, polysiloxane, polyurea, polyurethane, polyester or cellulose. Polyacrylate and polymethacrylate are preferred.
 - The dyestuffs, in particular the azo dyestuffs of the formula (I), are bonded covalently to these polymer matrices, as a rule via a spacer. For example, X^1 (or X^2) then represents such a spacer, in particular in the meaning $X^1 (Q^1) T^1 S^1$.

 $X^{1'}$

has the abovementioned meaning,

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Q¹ represents -O-, -S-, -(N-R⁵)-, -C(R⁶R⁷)-, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁸)-, -(CNR⁸-NR⁵)-, -(CH₂)_p-, p- or m-C₆H₄- or a divalent radical of the formula

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i

 T^{ι}

represents an integer from 0 to 4, where for i > 1 the individual Q^{I} can have different meanings,

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represents -(CH₂)_p-, where the chain can be interrupted by -O-, -NR⁹- or -OSiR¹⁰₂O-,

20 S¹

represents a direct bond, -O-, -S- or -NR9-,

p

represents an integer from 2 to 12, preferably 2 to 8, in particular 2 to 4,

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 \mathbb{R}^9

represents hydrogen, methyl, ethyl or propyl,

 R^{10}

represents methyl or ethyl and

R⁵ to R⁸

have the abovementioned meaning.

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Preferred dyestuff monomers for polyacrylates or -methacrylates then have the formula (II)

5

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wherein

15 R

represents hydrogen or methyl and

the other radicals have the abovementioned meaning.

Particularly suitable dyestuff monomers are those of the following formula (IIa)

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$$\begin{array}{c}
O \\
R
\end{array}$$

$$\begin{array}{c}
O \\
S^{1} - T^{1} \\
(Q^{1})_{i}
\end{array}$$

$$\begin{array}{c}
X^{1} \\
N^{2} \\
N
\end{array}$$

$$\begin{array}{c}
(R^{1})_{m} \\
X^{3}
\end{array}$$
(IIa),

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wherein

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X³ denotes CN and all other known electron-attracting substituents, and R¹ is then preferably also CN,

and the radicals R, S¹, T¹, Q¹, X¹ and R² and i, m and n have the abovementioned meaning.

The polymeric or oligomeric organic, amorphous material according to the invention can carry, in addition to the dyestuffs, for example of the formula (I), groupings having form anisotropy. These are also bonded covalently to the polymer matrices, as a rule via a spacer.

Groupings having form anisotropy have, for example, the structure of the formula (III)

wherein Z represents a radical of the formula

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$$(\mathbb{R}^{12}), \qquad \text{or}$$

wherein

A represents O, S or N-C₁- to C₄-alkyl,

30 X^3 represents $-X^{3'}-(Q^2)_j-T^2-S^2$ -,

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X⁴ represents X⁴-R¹³,

- $X^{3'}$ and $X^{4'}$ independently of one another represent a direct bond, -O-, -S-, -(N-R⁵)-, -C(R⁶R⁷)-, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁸)- or -(CNR⁸-NR⁵)-,
- R⁵, R⁸ and R¹³ independently of one another represent hydrogen, C₁- to C₂₀-alkyl, C₃to C₁₀-cycloalkyl, C₂- to C₂₀-alkenyl, C₆- to C₁₀-aryl, C₁- to C₂₀-alkyl(C=O)-, C₃- to C₁₀-cycloalkyl-(C=O)-, C₂- to C₂₀-alkenyl-(C=O)-, C₆to C₁₀-aryl-(C=O)-, C₁- to C₂₀-alkyl-(SO₂)-, C₃- to C₁₀-cycloalkyl(SO₂)-, C₂- to C₂₀-alkenyl-(SO₂)- or C₆- to C₁₀-aryl-(SO₂)- or
- X⁴-R¹³ can represent hydrogen, halogen, cyano, nitro, CF₃ or CCl₃,
- R^6 and R^7 independently of one another represent hydrogen, halogen, $C_1\text{--}$ to $C_{20}\text{--}$ alkyl, $C_1\text{--}$ to $C_{20}\text{--}$ alkoxy, $C_3\text{--}$ to $C_{10}\text{--}$ cycloalkyl, $C_2\text{--}$ to $C_{20}\text{--}$ alkenyl or $C_6\text{--}$ to $C_{10}\text{--}$ aryl,
- 20 Y represents a single bond, -COO-, -OCO-, -CONH-, -NHCO-, -CON(CH₃)-, -N(CH₃)CO-, -O-, -NH- or -N(CH₃)-,
- R¹¹, R¹², R¹⁵ independently of one another represent hydrogen, halogen, cyano, nitro, C₁- to C₂₀-alkyl, C₁- to C₂₀-alkoxy, phenoxy, C₃- to C₁₀-cycloalkyl, C₂- to C₂₀-alkenyl or C₆- to C₁₀-aryl, C₁- to C₂₀-alkyl-(C=O)-, C₆- to C₁₀-aryl-(C=O)-, C₁- to C₂₀-alkyl-(SO₂)-, C₁- to C₂₀-alkyl-(C=O)-NH-, C₆- to C₁₀-aryl-(C=O)-NH-, C₁- to C₂₀-alkyl-O-(C=O)-, C₁- to C₂₀-alkyl-NH-(C=O)- or C₆- to C₁₀-aryl-NH-(C=O)-,

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q, r and s independently of one another represent an integer from 0 to 4, preferably 0 to 2,

5 Q² represents -O-, -S-, -(N-R⁵)-, -C(R⁶R⁷)-, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁸)-, -(CNR⁸-NR⁵)-, -(CH₂)_p-, p- or m-C₆H₄- or a divalent radical of the formula

j represents an integer from 0 to 4, where for j > 1 the individual Q^1 can have different meanings,

T² represents -(CH₂)_p-, where the chain can be interrupted by -O-, -NR⁹- or -OSiR¹⁰₂O-,

S² represents a direct bond, -O-, -S- or -NR⁹-,

p represents an integer from 2 to 12, preferably 2 to 8, in particular 2 to 4,

R° represents hydrogen, methyl, ethyl or propyl and

R¹⁰ represents methyl or ethyl.

Monomers with such groupings having form anisotropy which are preferred for polyacrylates or -methacrylates then have the formula (IV)

$$\int_{R}^{Q} S^{2-T^{2}} (Q^{2})_{j}^{X^{3}} (R^{11})_{q}$$
 (IV),

wherein

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10 R represents hydrogen or methyl and

the other radicals have the abovementioned meaning.

Particularly preferred monomers having form anisotropy of the formula (IV) are, for example:

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The alkyl, cycloalkyl, alkenyl and aryl radicals can in turn be substituted by up to 3 radicals from the series consisting of halogen, cyano, nitro, C_1 - to C_{20} -alkyl, C_1 - to C_{20} -alkoxy, C_3 - to C_{10} -cycloalkyl, C_2 - to C_{20} -alkenyl or C_6 - to C_{10} -aryl, and the alkyl and alkenyl radicals can be straight-chain or branched.

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Halogen is to be understood as meaning fluorine, chlorine, bromine and iodine, in particular fluorine and chlorine.

In addition to these functional units, the oligo- or polymers according to the invention can also comprise units which serve chiefly to lower the percentage content of functional units, in particular dyestuff units. In addition to this task, they can also be responsible for other properties of the oligo- or polymers, e.g. the glass transition temperature, liquid crystallinity, film formation properties etc.

For polyacrylates or -methacrylates, such monomers are acrylic or methacrylic acid esters of the formula (V)

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wherein

R represents hydrogen or methyl and

25 R¹⁴

represents optionally branched C_{1} - to C_{20} -alkyl or represents a radical containing at least one further acrylic unit.

However, other copolymers can also be present.

Polyacrylates and polymethacrylates according to the invention then comprise as recurring units preferably those of formula (VI), preferably those of formulae (VI) and (VII) or of formulae (VI) and (VIII) or those of formulae (VI), (VII) and (VIII)

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wherein the radicals have the abovementioned meaning.

It is also possible for there to be present several of the recurring units of formula (VI) and/or of the recurring units of the formulae (VII) and/or (VIII).

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The ratio of amounts between VI, VII and VIII can be as desired. Preferably, the concentration of VI is between 0.1 and 100%, based on the particular mixture, depending on the absorption coefficient of VI. The ratio between VI and VII is between 100:0 and 1:99, preferably between 100:0 and 30:70, especially preferably between 100:0 and 50:50.

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The polymers and oligomers according to the invention preferably have glass transition temperatures T_g of at least 40°C. The glass transition temperature can be determined, for example, by the method of B. Vollmer, Grundriß der Makromolekularen Chemie [Basic Principles of Macromolecular Chemistry], p. 406-410, Springer-Verlag, Heidelberg 1962.

The polymers and oligomers according to the invention have a molecular weight, determined as the weight-average, of 5,000 to 2,000,000, preferably 8,000 to 1,500,000, determined by gel permeation chromatography (calibrated with polystyrene).

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As a result of the structure of the polymers and oligomers, the intermolecular interactions of the structural elements of the formula (VI) with one another or of the formulae (VI) and (VII) with one another are established such that the formation of liquid crystal states of order is suppressed and optically isotropic, transparent non-scattering films, foils, sheets or parallelepipeds can be produced. On the other hand, the intermolecular interactions are nevertheless strong enough for a photochemically induced, cooperative, directed reorientation process of the photochromic and non-photochromic side groups to be effected on irradiation with light.

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Preferably, the interaction forces which occur between the side groups of the recurring units of the formula (VI) or between those of the formulae (VI) and (VII) are sufficient for the photo-induced change in configuration of the side groups of the formula (VI) to cause a reorientation in the same direction - so-called cooperative - of the other side groups ((VI) and/or (VII)).

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Extremely high values of the optical anisotropy can be induced in the optically isotropic amorphous photochromic polymers (Δn to 0.4).

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By the influence of actinic light, states of order are generated and modified in the polymers or oligomers and the optical properties are thus modulated.

The light used is polarized light, the wavelength of which lies in the range of the absorption band, preferably in the range of the long wavelength $n-\pi^*$ band of the recurring units of the formula (VI).

The polymers and oligomers can be prepared by processes known from the literature, for example from DD 276 297, DE-A 3 808 430, Makromolekulare Chemie 187, 1327-1334 (1984), SU 887 574, Europ. Polym. 18, 561 (1982) and Liq. Cryst. 2, 195 (1987).

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Another method of preparing the recording material or the polymer according to the invention comprises a process in which at least one monomer is polymerized without a further solvent, it preferably being polymerized by free radicals, and the polymerization particularly preferably being initiated by free-radical initiators and/or UV light and/or thermally.

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The reaction is carried out at temperatures between 20°C and 200°C, preferably between 40°C and 150°C, particularly preferably 50°C and 100°C, and especially preferably at about 60°C.

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In a particular embodiment, AIBN is used as a free-radical initiator.

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It has often proved favourable to co-employ another, preferably liquid monomer. This is understood as meaning monomers which are liquid at the reaction temperatures and are preferably olefinically unsaturated monomers, particularly preferably based on acrylic acid and methacrylic acid, especially preferably methyl methacrylate.

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The content of monomers of the formula (II) in the copolymers is preferably 0.1 to 99.9 wt.%, particularly preferably 0.1 to 50 wt.%, especially preferably 0.1 to 5 wt.%, and in the most favourable case 0.5 to 2 wt.%.

Particularly suitable materials comprise at least one of the polymers of the formula XIII to XX

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The invention also provides a preparation process for suitable monomers analogous to the formula under example 1.2.

The process is characterized in that terephthalic acid dichloride is stirred with an aminoazobenzene or a substituted aminoazobenzene in at least equimolar amounts at a temperature between room temperature and 110°C in an inert solvent, e.g. dioxane, the unsymmetric partial amide being formed quantitatively, which can be filtered off with suction immediately. The expert sees immediately that substituted aminoazobenzenes are of course also suitable. The product is optionally washed again with the solvent. An optionally substituted 4-(carboxychloride)-benzoic acid (4'-phenylazo)-benzamide is then obtained.

The invention also provides an unsymmetric terephthalamide prepared in such a way, and the optionally substituted unsymmetric terephthalamide and the acrylate or methacrylate which can be prepared therefrom according to example 1.2.

The invention also provides a monomer according to example 1.3 of the present application, which can of course also be prepared in substituted form.

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Films, foils, sheets and parallelepipeds are produced without expensive orientation processes utilizing external fields and/or surface effects being necessary. They can be applied to substrates by spin-coating, dipping, pouring or other coating processes which are technologically easy to conduct, introduced between two transparent sheets by pressing or flowing in, or prepared simply as a self-supporting material by casting or extrusion. Such films, foils, sheets and parallelepipeds can be produced by sudden cooling, i.e. by a cooling rate of > 100 K/min, or also from liquid crystal polymers or oligomers which contain structural elements in the sense described by rapid stripping off of the solvent.

Preferred preparation processes for recording materials for binary and/or multibit and/or volume data storage comprise a step in which the storage medium is applied by spin-coating.

Preferred production processes for holographic volume storage systems comprise a step according to a conventional injection moulding process in the range up to 300°C, preferably up to 220°C, particularly preferably 180°C.

The layer thickness is ≥ 0.1 mm, preferably ≥ 0.5 mm, particularly preferably ≥ 1 mm. A particularly preferred preparation process for layers in the millimetre range is the injection moulding process. In this, the polymer melt is forced through a nozzle into a shaping holder, from which it can be removed after cooling.

A STATE AND AS

Examples:

Example 1 Preparation of the monomers

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85.9 g 4-aminodiphenyl ether in 200 ml dioxane are added to a solution of 125 g 4-(2-methacryloyloxy)-ethoxy-benzoic acid chloride in 200 ml dioxane, the mixture is stirred for 2 h and the product is precipitated by pouring the solution into 2 l water. The precipitate is filtered off with suction, dried and purified by recrystallization twice from isopropanol. The yield is 80% of theory. M.p. = 123°C

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59 g 4-aminoazobenzene in 400 ml dioxane are added to a solution of 203 g terephthalic acid dichloride in 1,000 ml dioxane and the mixture is stirred for 1 h. The precipitate is filtered off with suction, rinsed with dioxane and dried under a high vacuum at 110°C for 8 h. The yield of 4-(carboxychloride)-benzoic acid (4'-phenylazo)-benzamide is 92.8 g. M.p. = 123°C

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Elemental analysis: C₂₀H₁₄ClN₃O₂ (363.81)

Calc.: C66.03; H3.88; C19.75; N11.55 Found: C66.10; H4.00: C19.70; N11.70

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26 g 2-hydroxyethyl methacrylate in 25 ml pyridine are added to a solution of 36.4 g of this substance in 200 ml DMF, the mixture is stirred at 100°C for 2 h and the product is precipitated by pouring the solution into 1 l water. The precipitate is filtered off with suction, dried and purified by chromatography in toluene/ethyl acetate (1:1) on silica gel. The yield is 40% of theory.

Elemental analysis: C₂₆H₂₃N₃O₅ (457.49)

Calc.: C68.26; H5.07; N9.18. Found: C68.00; H5.10; N9.20.

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4-Hydroxy-3-bromo-4'-cyanoazobenzene

18.8 g 4-cyanoaniline are heated at 60°C in 100 ml water. To dissolve the solid, 10 ml conc. hydrochloric acid are added. A further 115 ml hydrochloric acid are then added, the temperature of the solution is brought to 0°C, 52 g nitrosylsulfuric acid are slowly added dropwise and the mixture is stirred for 1 h. This solution is slowly added dropwise to a solution of 27.5 g 2-bromophenol, 100 ml methanol and 100 ml water at 10°C. A pH of 6 - 6.5 is maintained by simultaneous addition of 10% sodium hydroxide solution. When the addition is complete, the stirred mass is allowed to react for a further hour and the precipitate is filtered off with suction. The product is then washed with water and dried. The yield of 4-hydroxy-3-bromo-4'-cyanoazobenzene is 68 g.

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4-(2-Hydroxyethyloxy)-3-bromo-4'-cyanoazobenzene

65 g 4-Hydroxy-3-bromo-4'-cyanoazobenzene are dissolved in 200 ml 4-methyl-2-pentanone at approx. 120°C. 40 g 2-bromoethanol, 45 g potassium carbonate and 0.5 g potassium iodide are added to this solution and the mixture is stirred at 120°C for 24 h. The product is taken up in chloroform and washed with water. After drying with magnesium sulfate, the solvent is evaporated off on a rotary evaporator and the residue is purified by chromatography on silica gel in toluene/ethyl acetate (4/1).

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The yield of 4-(2-hydroxyethyloxy)-3-bromo-4'-cyanoazobenzene is 30.7 g. M.p. = 153° C; $_{\text{max}} = 365 \text{ nm (DMF)}$

15 Elemental analysis: $C_{15}H_{12}BrN_3O_2$ (346.19)

Calc.: C52.04; H3.49; Br23.08; N12.14. Found: C52.30; H3.30; Br22.90; N12.10.

- 4-(2-Hydroxyethyloxy)-3,4'-dicyanoazobenzene

15 g (4-(2-hydroxyethyloxy)-3-bromo-4'-cyanoazobenzene are dissolved in 25 ml hot DMF. 4.3 g copper cyanide are then added and the reaction mixture is stirred at 140°C for 5-6 hours. This solution is added to approx. 500 ml of a 13% aqueous ammonia solution and the precipitate is filtered off. The precipitate is dissolved in hot dioxane. The solution is filtered off from the undissolved residue, the dioxane is evaporated off on a rotary evaporator and the product is purified by chromatography on silica gel in toluenc/THF (1/2) and recrystallized once from ethanol. The yield of 4-(2-hydroxyethyloxy)-3,4'-dicyanoazobenzene is 4.5 g. M.p. = 138°C; max = 356 nm (DMF)

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4-(2-Methacryloyloxyethyloxy)-3,4'-dicyanoazobenzene

5.8 g (4-(2-hydroxyethyloxy)-3,4'-dicyanoazobenzene are dissolved in 15 ml dioxane at 65°C; 2.4 g triethylamine are added. A solution of 3.1 g methacrylic acid chloride in 3 ml dioxane is added and the reaction mixture is stirred at 65°C for 2 h and then introduced into 350 ml ice-water. The precipitate is filtered off with suction and dried. Purification is carried out by chromatography on silica gel in toluene/THF (1/2). The yield of 4-(2-methacryloyloxyethyloxy)-3,4'-dicyanoazobenzene is 5.9 g. M.p. = 110°C; max = 352 nm (DMF)

Example 2 Preparation of the polymers

3 g monomer 1.2 and 0.15 g 2,2'-azoisobutyric acid dinitrile are stirred in 30 ml DMF in an argon atmosphere at 70°C for 24 h. The polymer is precipitated by pouring the solution into 200 ml water and purified by boiling up in methanol.

Example 3:

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A glass substrate 1 mm thick is provided with a thin layer of the polymer (see above, example 3). This is effected with the aid of the spin-coating technique. In this procedure, the polymer is dissolved in tetrahydrofuran at a concentration of 50 g/l and the solution is dripped on to the substrate, which is rotating at a speed of rotation

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of 2,000 min⁻¹. The polymer film formed typically has a thickness of 200 nm. Residues of the solvent are removed from the film by storage of the coated glass support for 2 h at 60°C in a vacuum oven.

The sample prepared in this way is irradiated from the polymer side with polarized laser light incident perpendicularly (writing operation). An argon ion laser (Continuum) at the wavelength of 514 nm is used as the light source. The intensity of this so-called writing laser is 500 mW/cm². trans-cis-trans Isomerization cycles are induced in the side group molecules of the polymer, which leads to a build up of a net orientation of the molecules away from the direction of polarization of the Ar laser. These molecular dynamics manifest themselves macroscopically in a developing birefringence $\Delta n=n_y-n_x$ in the polymer film plane. The refractive index in the direction of polarization of the laser light (n_x) decreases during this process, while the refractive index perpendicular to the direction of polarization (n_y) increases. With the stated exposure parameters, the dynamics proceed in the range of minutes.

The course with respect to time of the induced birefringence at a wavelength of 633 nm is determined experimentally with a helium-neon laser (typical intensity: 10 mW/cm^2). This operation is called reading the birefringence. The light of this so-called reading laser incident on the polymer layer occupies a fixed angle of between 15° and 35° to the perpendicular of the layer. The reading and writing light overlap on the polymer layer. The direction of polarization of the reading light occupies an angle in the polymer film plane of 45° to the polarization of the writing light. It is rotated on passing through the polymer layer if the layer is birefringent. This rotation is accompanied by an increase in the reading light intensity I_s after an analyser, which is in the beam path after the sample and allows through light perpendicular to the original direction of polarization. The intensity I_p decreases to the same extent as I_s increases. I_p is defined as the transmitted intensity after an analyser which is positioned likewise but which selects the original direction of

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polarization of the reading laser. The two contents of the direction of polarization parallel and perpendicular to the original direction are detected by experiment separately via a polarizing beam divider and with the aid of two Si photodiodes. The birefringence Δn is calculated from the intensities measured via the following relationship:

$$\Delta n = \frac{\lambda}{\pi d} \arcsin \sqrt{\frac{I_s}{I_s + I_p}}$$

wherein d designates the thickness of the polymer layer and $\lambda = 633$ nm, the light wavelength of the reading laser. In this formula, it is assumed by approximation that reading is carried out perpendicularly to the polymer layer.

Fig. 1 shows the monotonous increase with respect to time of the birefringence Δn of a layer of the polymer from example 3 during writing/deletion experiments. A high birefringence value indicates a high anisotropy in the orientation distribution of the chromophor in the film plane. After exposure of the sample to the light of the writing laser for 2 minutes, the first writing operation is concluded. The resulting phase shift $\Delta \phi = 2\pi \Delta n d / \lambda$ during this and subsequent writing operations does not exceed the value $\Delta \phi = \pi$. The birefringence n of the polymer layer has reached virtually a maximum value of $\Delta n = 0.213 \pm 0.002$ after 2 min.

 Δn is deleted by rotating the direction of polarization of the writing light through 90°. This deletion operation is concluded as soon as: $\Delta n = 0$. This is equivalent to a value $I_s = 0$, which is detected via a diode. The deletion is effected here significantly faster than the writing.

Further writing/deletion operations follow these first directly in the same pattern, and the diode signals are recorded and the birefringence calculated. It can be seen in fig. 1 that the build up of the birefringence in the second and all subsequent writing operations is comparable in speed and level in the context of measurement accuracy.

The polymer therefore does not bleach out, which would be deduced from a successive decrease in the birefringence.

Patent claims

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- Optical recording material for binary and/or multibit and/or volume data storage, comprising at least one polymeric and/or oligomeric azo dyestuff which changes its spatial arrangement on irradiation with polarized electromagnetic radiation, and optionally at least one grouping having form anisotropy, characterized in that
 - the absorption maximum of the dyestuff is at least 30 nm, preferably 40 nm, different from 400 nm and
 - at 400 nm the dyestuff reaches an optical density of not more than 60% of its absorption maximum and
 - there is the capacity for being rewritten on by changing the state of polarization of the actinic light, an intensity of at least 80% of the original value being achieved after a deletion/rewriting cycle, and
- wherein at 400 nm, under otherwise identical conditions, the writing operation proceeds no more slowly than at 500 nm, and birefringence values induced here do not differ from those birefringence values induced at 500 nm by more than 10%.
- 2. Recording material according to claim 1, characterized in that the absorption maximum (AM) of the dyestuff is less than 370 nm, preferably 360 nm.
 - 3. Recording material according to claim 1, characterized in that the absorption maximum of the dyestuff is greater than 450 nm.

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- 4. Recording material according to claim 1, characterized in that it comprises a copolymer which comprises at least one component of which the AM is greater than 450 nm, and at least one component of which the AM is less than 360 nm.
- 5. Recording material according to one or more of claims 1 to 4, characterized in that in the solid state at a thickness of 250 nm it has an optical density of ≤ 1, preferably less than or equal to 0.5, especially preferably less than or equal to 0.3, at a wavelength in a wavelength range from 380 to 420 nm, preferably 390 to 410 nm, especially preferably 395 to 405 nm.
- 6. Recording material according to one or more of claims 1 to 5, characterized in that the electromagnetic radiation is light in the laser wavelength range of preferably between 380 to 420 nm, particularly preferably between 390 and 410 nm, especially preferably between 395 and 405 nm.
- 7. Recording material according to any one of claims 1 to 6, characterized in that the chemically bonded dyestuff corresponds to the formula (I):

$$X^{1} \qquad (R^{1})_{m} \qquad (I),$$

wherein

R¹ and R² independently of one another represent hydrogen or a nonionic substituent and

m and n independently of one another represent an integer from 0 to 4, preferably 0 to 2, where

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- X^1 and X^2 denote $X^{1'}-R^3$ or $X^{2'}-R^4$, and
- $X^{1'}$ and $X^{2'}$ represent a direct bond, -O-, -S-, -(N-R⁵)-, -C(R⁶R⁷)-, -(C=O)-, -(CO-O)-, -(CO-NR⁵)-, -(SO₂-O)-, -(SO₂-O)-, -(SO₂-NR⁵)-, -(C=NR⁸)- or -(CNR⁸-NR⁵)-,
 - R³, R⁴, R⁵ and R³ independently of one another represent hydrogen, C_1 to C_{20} -alkyl, C_3 to C_{10} -cycloalkyl, C_2 to C_{20} -alkenyl, C_6 to C_{10} -aryl, C_1 to C_{20} -alkyl-(C=O)-, C_3 to C_{10} -cycloalkyl-(C=O)-, C_2 to C_{20} -alkenyl-(C=O)-, C_6 to C_{10} -aryl-(C=O)-, C_1 to C_{20} -alkyl-(SO₂)-, C_3 to C_{10} -cycloalkyl-(SO₂)-, C_2 to C_{20} -alkenyl-(SO₂)- or C_6 to C_{10} -aryl-(SO₂)- or
- 15 X''-R³ and X^{2'}-R⁴ can represent hydrogen, halogen, cyano, nitro, CF₃ or CCl₃,
 - R^6 and R^7 independently of one another represent hydrogen, halogen, C_1 to C_{20} -alkyl, C_1 to C_{20} -alkoxy, C_3 to C_{10} -cycloalkyl, C_2 to C_{20} -alkenyl or C_6 to C_{10} -aryl.

wherein the sensitivity of the dyestuff to actinic light after the induced birefringence has been written in is retained at a final value,

- and this sensitivity is at least 5%, preferably 10%, particularly preferably 15%, especially preferably 20% of the original value when the longitudinal axis of the molecule lies perpendicular to the direction of polarization of the actinic light.
- 8. Recording material according to any one of claims 1 to 7, characterized in that at least one monomer of the formula (II)

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wherein

R represents hydrogen or methyl and

the other radicals have the abovementioned meaning,

has been used in the preparation.

9. Recording material according to any one of claims 1 to 8, characterized in that it comprises at least one of the polymers of the formula XIII to XX

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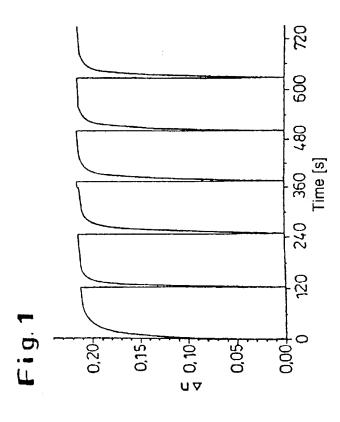
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wherein the compositions in the co- and terpolymers can vary, provided that x + y adds up to 100 mol%, or x + y + z adds up to 100 mol%.

- 10. Recording material according to claim 9, characterized in that p is between 10 and 1,000 and/or in the case of the copolymers x: y is between 10:90 and 90:10, preferably between 30:70, particularly preferably between 40:60 and 60:40, and especially preferably 50:50, and/or in the case of terpolymers x + y is greater than 10 mol%, preferably greater than 20 mol%, particularly preferably greater than 30 mol%.
 - 11. Storage system, characterized in that it comprises a recording material according to any of claims 1 to 10.
 - 12. Storage system according to claim 11, characterized in that the recording material comprises one or more unsupported objects of any desired shape, preferably an unsupported flat structure, particularly preferably an unsupported film, a multi-layer build-up preferably comprising at least one substrate layer.
- 20 13. Storage system according to claim 11, characterized in that it also additionally comprises a reflection layer.
 - 14. Process for the production of the storage system according to at least one of claims 11 or 12, or 13, characterized in that it comprises a step in which the storage medium is applied by spin-coating.



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INTERNATIONALE ANMELDUNG VERÖFFENTLICHT NACH DEM VERTRAG ÜBER DIE
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(21) Internationales Aktenzeichen: PCT/EPC (22) Internationales Anmeldedatum: 17. März 2000 ((30) Prioritätsdaten: 199 14 325.0 30. März 1999 (30.03.99) (71) Anmelder (für alle Bestimmungsstaaten ausser US): AKTIENGESELLSCHAFT [DE/DE]; D-51368 Le (DE). (72) Erfinder; und (75) Erfinder/Anmelder (nur für US): BERNETH, Horst Erfurter Strasse 1, D-51373 Leverkusen (DE). BIEI Thomas [DE/DE]; Am Pützchen 25, D-51519 (DE). HAGEN, Rainer [DE/DE]; Felix-von- 35, D-51375 Leverkusen (DE). KOSTROMINE [RU/DE]; Katharinenstrasse 28, D-53913 Swisttal (74) Gemeinsamer Vertreter: BAYER AKT SELLSCHAFT; D-51368 Leverkusen (DE).	BAYE everkuse [DE/DE RINGE Odenth Roll-S, Sergu	BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TI, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW. ARIPO Patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), eurasisches Patent (AM, AZ, BY, KG, KZ, MD, RU, TI, TM), europäisches Patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI Patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Veröffentlicht Mit internationalem Recherchenbericht.		
(54) Title: ERASABLE OPTICAL RECORDING MATERIAL FOR BLUE LASERS (54) Bezeichnung: WIEDERBESCHREIBBARES OPTISCHES AUFZEICHNUNGSMATERIAL FÜR BLAUE LASER (57) Abstract The invention relates to novel optical recording materials for storing binary and/or multi-bit and/or volume data, their production and their use as storage material at wavelengths around 400 nm. (57) Zusammenfassung Es werden neue optische Aufzeichnungsmaterialien für die binäre und/oder Multibit und/oder Volumendatenspeicherung, deren Herstellung und Verwendung als Speichermaterial bei Wellenlängen um 400 nm beschrieben.				

COMBINED DECLARATION AND POWER OF ATTORNEY

COMBINED DISCHARITION AND TO WAR OF THE FOLLOW

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought

on the invention entitled

ERASABLE OPTICAL RECORDING MATERIAL FOR BLUE LASERS

the specification of which is attached hereto,

or was filed on March 17, 2000

as a PCT Application Serial No. PCT/EP00/02385

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s), the priority(ies) of which is/are to be claimed:

199 14 325.0 (Number)

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Germany (Country)

March 30, 1999 (Month/Day/Year Filed)

I hereby claim the benefit under Title 35, United States Code, \$120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, \$112, I acknowledge the duty to disclose the material information as defined in Title 37, Code of Federal Regulations, \$1.56 which occured between the filing date of the prior application and the national or PCT international filing date of this application:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Le A 33 661-US

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JOSEPH C. GIL, Patent Office Registration Number 26,602 ARON PREIS, Patent Office Registration Number 29,426 LYNDANNE M. WHALEN, Patent Office Registration Number 29,457 THOMAS W. ROY, Patent Office Registration Number 29,582 RICHARD E. L. HENDERSON, Patent Office Registration Number 31,619 GODFRIED R. AKORLI, Patent Office Registration Number 28,779 N. DENISE BROWN, Patent Office Registration Number 36,097 NOLAND J. CHEUNG, Patent Office Registration Number 39,138 DIDERICO VAN EYL, Patent Office Registration Number 38,641 CAROLYN M. SLOANE, Patent Office Registration Number 44,339 JAMES R. FRANKS, Patent Office Registration Number 42,552

JACKIE ANN ZURCHER, Patent Office Registration Number 42,251
RAYMOND J. HARMUTH, Patent Office Registration Number 33,896

all of Bayer Corporation, Pittsburgh, Pennsylvania 15205-9741 Send Correspondence To: Direct Telephone Calls To: Patent Department Bayer Corporation (412) 777-2349 100 Bayer Road Pittsburgh, Pennsylvania 15205-9741 DATE INVENTOR'S, SIGNATURE FULL NAME OF SOLE OR FIRST INVENTOR 21-07-10 Horst Rerneth CITIZENSHIP RESIDENCE German D 51373 Leverkusen, Germany POST OFFICE ADDRESS c/o Bayer Aktiengesellschaft, D 51368 Leverkusen, Germany DATE INVENTOR'S SIGNATURE FULL NAME OF SECOND INVENTOR 2001-07-31 Thomas Bieringer CITIZENSHIP RESIDENCE German D 51519 Odenthal, Germany POST OFFICE ADDRESS D 51368 Leverkusen, Germany c/o Bayer Aktiengesellschaft, DATE 2001 - 07-41 INVENTOR'S SIGNATURE FULL NAME OF THIRD INVENTOR Rainer Hagen CITIZENSHIP RESIDENCE German D 51373 Leverkusen, Germany POST OFFICE ADDRESS c/o Bayer Aktiengesellschaft, D 51368 Leverkusen, Germany INVENTOR'S SIGNATURE DATE FULL NAME OF FOURTH INVENTOR 1500111 200107_30 Serguei Kostromine CIGUES CITIZENSHIP RESIDENCE DEX Russian D 53913 Swisttal, Germany POST OFFICE ADDRESS c/o Bayer Aktiengesellschaft, D 51368 Leverkusen, Germany DATE INVENTOR'S SIGNATURE FULL NAME OF FIFTH INVENTOR CITIZENSHIP RESIDENCE POST OFFICE ADDRESS DATE INVENTOR'S SIGNATURE FULL NAME OF SIXTH INVENTOR CITIZENSHIP RESIDENCE POST OFFICE ADDRESS DATE INVENTOR'S SIGNATURE FULL NAME OF SEVENTH INVENTOR CITIZENSHIP RESIDENCE POST OFFICE ADDRESS